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**14. ABSTRACT**

The project increased the highest power obtained for a diamond Raman laser by 15 times and up to 0.4 kW. A continuous wave diamond laser was demonstrated that generated 108 W at 1240 nm from a 320 W Nd:YAG pump laser. A modified design generating 380 W was demonstrated using a 630 W Ybdoped fiber laser system. In each case the performance was unsaturated and limited by the available pump power. The efficiencies and brightness achieved are found to be higher than expected by current theories for thermal effects in diamond. The project also developed a diamond laser capable of providing large increases in beam quality (from M2 = 3.5 to M2 < 1.1) and brightness (by 1.7 times), features that were used to demonstrate a pulsed eye-safe (1.5 m) laser based on a Nd:YAG pump with an unprecedented brightness of 540 MW/cm2/sr.

What is/are the significance of the findings:

The results are significant in showing that diamond Raman laser technology is an efficient wavelength converter for conventional high power laser technologies including Nd doped lasers and Yb-doped fiber lasers. Diamond’s power handling capability now exceeds the highest achieved for any other Raman crystal by a factor of 22 and is competitive with Raman fibers. In addition, it is highly promising for brightness enhancement of beams and for beam combining of incoherent beams based on the technique of Raman beam combination. Compared to other nonlinear conversion schemes (such as harmonic generation in crystals and Raman fibers), diamond has important advantages of large wavelength Stokes shift, wavelength range (from UV to mid-IR) and suitability for conversion a variety of temporal domains (ultrafast to cw). It is thus found to be an outstanding nonlinear optical material in terms of many important categories of high power performance.

The project raises several questions for further research.

As power scaling is a major motivation for studying diamond, one of the most important questions pertains to the extent to which laser power can be scaled before thermal effects become evident. The mechanism for heat deposition and its spatial dependence are found to be poorly understood. Thus further experiments with accompanying theoretical development are crucial to the development of accurate models.

**15. SUBJECT TERMS**

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**Final Report for AOARD Grant 124055**
**"High average power Raman conversion in diamond:**
**'Eyesafe' output and fiber laser conversion"**

**19 June 2015**

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**Abstract:**
Synthetic diamond is an interesting nonlinear optical material with potential for bringing about radical enhancements in Raman laser average power, brightness and wavelength range. This report details several aspects related to enhanced Raman laser performance including power scaling, high brightness generation and application to fiber laser technology. Major results include: Demonstration of a continuous wave diamond laser with 380 W output power – an increase of 15 times and 22 times than higher than previously reported for any crystalline Raman laser. Demonstration of a high brightness pulsed second-Stokes laser suitable for high-pulse-rate eye-safe laser applications. Development of a novel analytical model for optimization of continuous wave diamond Raman lasers in the non-thermal regime. Demonstration of efficient (up 60%) Raman conversion of cw fiber lasers of power up to 630 W. Several further findings are made in relation to other nonlinear diamond effects and diamond damage. Overall, the results show that diamond is a practical and efficient wavelength and brightness converter that can be applied to a variety of pump laser technologies at average input powers above 0.6 kW and with excellent prospects for extension beyond a kilowatt.

**Introduction:**

Ever since the discovery of the laser, there has been a large research effort to develop laser technologies of increasing power and with diverse performance characteristics. Presently the highest power systems exceed hundreds of kilowatts and are based on gas discharges (e.g. $CO_2$ lasers at 10.6 μm), or chemical lasers that rely on fast-flow chemical reactions of gases (COIL and HF/DF lasers). In the last 5-10 years, advances in laser materials and high power (laser diode) pump technology have led to the development of more practical solid-state alternatives. Laser rod technologies based on rare-earths such as Nd, Yb or Tm have been adapted to withstand the extreme power handling requirements by increasing waste heat dissipation through the use of geometries such as slabs, thin-disk and fiber lasers. The need for systems that are simultaneously lightweight, small footprint, non-polluting, and efficient, has put increasing reliance on solid-state approaches. The major challenges presently are to increase laser power of these systems while maintaining high beam quality and with diversified wavelength range.

Raman lasers comprise a promising route to overcome these challenges. As well as providing a method for shifting the wavelength, Raman lasers are capable of transferring power from multiple input modes into a single output spatial mode. However, until recently Raman lasers have been restricted to relatively low power (< 30 W) due to the substantial heat deposition in the Raman material as a result of phonon field decay. A further issue is the requirement for highly coherent pumps to drive the relatively weak third-order nonlinear interaction.

Two major and very recent developments have enabled beam conversion at much higher power. Raman fiber lasers, in which the fiber waveguide simultaneously enhances the nonlinear interaction and mitigates thermal lensing, have exploited high-power Yb fiber laser technology at 1.08 μm to generate 1-1.3 kW at 1.12 μm.[1] Diamond Raman lasers comprise a second emerging technology with many major attractions. Diamond's thermal conductivity (approx. 2000 W/m/K) is two orders of magnitude higher than most optical materials and all other known viable laser materials. Coupled with a low thermal expansion coefficient, there are excellent prospects for developing very high power lasers with diffraction-limited beam quality ***in bulk at room temperature***, i.e. without resorting to more complex geometries (waveguide, slab or disk) or cryogenic cooling to mitigate thermal effects. The Raman shift of 1332 $cm^{-1}$ and working wavelength range of the material are much greater than fiber Raman technology. Wavelengths of high-power fiber Raman lasers are limited to approximately 1-2 μm by photo-darkening at short wavelengths and absorption bands of the silica constituents of the glass at long wavelengths. Diamond's large Raman shift (four times larger than glass fibers) provides access to more regions of the spectrum and can potentially span most of its very wide transmission band from 0.23 – 4 μm, and possibly in the applications rich and highly-prized wavelength range longer than 6 μm. The Raman gain coefficient is amongst the highest for Raman materials (approx 10 cm/GW at 1 μm), a key benefit for enabling efficient devices and relaxing the constraints on pump beam coherence.

Prior to this project, diamond Raman lasers (DRLs) had been investigated using crystalline Nd or Yb doped pumps with output powers up to 16 W for pulsed and cw lasers[2], and one report[3] of a laser producing two counter-propagating output beams with 12.5 W each. In order to take DRL technology to the next level of performance and assess its potential as a generic technology for high power beam conversion, the objectives of this 3-year project were to:

1. Increase DRL output power to the 100 W level;

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[1] L. Zhang, Chi Liu, Huawei Jiang, Yunfeng Qi, Bing He, Jun Zhou, *et al* Opt. Express **22**, 18483-18489. (2014)
[2] AOARD Report 104078, 7 Jan 2012
[3] Feve, J.-P. M., Shortoff, K. E., Bohn, M. J., & Brasseur, J. K. Opt. Express, **19**, 913–22, (2011).

2. Investigate high average power conversion of 1 µm laser technology to the 1.5 µm eye-safe region;
3. Adapt DRL converter technology to tunable fiber pump lasers;
4. Augment models to enable prediction of performance when cascading to higher Stokes orders and increasing power beyond the 100 W level.

These are aimed to enable a detailed assessment of the potential for DRLs to provide high power conversion of standard crystal and fiber laser technologies, understand factors limiting efficiency, and develop design rules for scaling towards kW power levels.

**Results and Discussion:**
The results/discussion is divided into five tasks aligned with the above four adjectives and an additional category describing significant additional outcomes from the project that were not anticipated in the original project design.

1) **Power scaling**
2) **Eye-safe (1.5 µm) DRLs**
3) **Fiber laser conversion**
4) **Model development**
5) **Additional outcomes**

For each, descriptions are given for the major findings, their significance to the field and future work that will be beneficial.

**1) Power scaling**
The first tranche of experiments were performed using quasi-cw pump lasers. The rapid thermal dissipation rate of diamond enables investigation of many steady-state high-power operational characteristics in short bursts of operation. Steady-state output is attained at a faster rate than in most other crystals by approximately 3 orders of magnitude (see inset figure) and well within the typical 0.1 – 0.3 ms pulse duration of quasi-cw lasers. Thus by using quasi-cw Nd:YAG technology we were able to readily investigate performance at pump powers up to 320 W for durations corresponding to at least 10 times the duration needed to establish steady-state thermal gradients. Models were used to optimize cavity design, pump polarization and coating specifications.

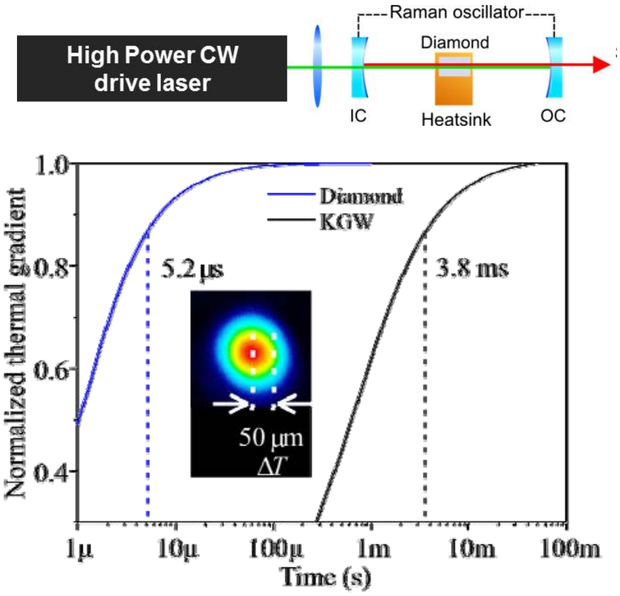


**Figure 1 Experimental concept and calculated time response for thermal gradients (50 µm waist radius).**

This approach yielded **108 W** of output power at the 1240 nm first-Stokes wavelength, which was an order of magnitude above previous cw and quasi-cw crystalline Raman lasers. The DRL operated with 34% conversion efficiency and excellent beam quality ($M^2$<1.1), and showed no evidence of thermal effects or damage to the diamond even at the highest power. These results, in concert with the models developed in Sec 4, demonstrated the capability for diamond to provide cw beam conversion at output power levels above 100 W, and foreshadowed applications for DRLs as compact and efficient frequency converters for high-power laser platforms such as narrow-linewidth fiber lasers and thin-disk lasers.

The second tranche of experiments used a 700 W fiber laser as the pump source, performed in collaboration with the Fraunhofer Institute of Applied Optics and Precision Engineering in Jena who had developed the high power pump laser. The higher power introduced a range of challenges including the development of approaches to isolate the fiber amplifier from back-reflected pump light returning from the DRL cavity. One approach used geometric isolation by applying tilt to the back-reflecting components of the DRL cavity - namely the input-coupler (IC), diamond, and output-coupler (OC) - such that the back-reflected pump beams were directed onto a beam-block. In order to minimize the degree of tilt applied to the cavity mirrors, a path length of approximately 15 m between the fiber amplifier and the DRL was used, and the tilt applied to the mirrors was less than 1 mrad. As this approach does not introduce additional components into the beam path, it offers simplicity and low-loss and does not degrade the pump beam quality. The second approach used a thin-plate polarizer and a quarter-wave plate in combination as isolation. This alternative to Faraday isolators allows co-linear alignment of the external cavity to a circularly-polarized pump beam but is susceptible to depolarization effects in the diamond due to stress-induced birefringence.

The circular polarization pump was the most successful. Steady-state operation (millisecond-pulses) at **381 W** of output power was observed at 1234 nm. The 61% conversion efficiency obtained is comparable to the record DRL fficiencies that have been obtained using ns-pulsed pumping. Fundamental mode output was obtained without saturation. The output power is 15 times higher than the previous power record for diamond[3] and 22 times higher than any other crystal Raman laser material. Calculations indicate that the diamond is operating well-beyond the expected stability limit and that the thermal lens in the diamond is much weaker than anticipated. This finding challenges the current understanding of optical phonon decay processes and heat generation in Raman crystals, a subject that will require further investigation. A more detailed knowledge of the spatial dependence of phonon transport and decay is crucial for predicting performance at higher powers.

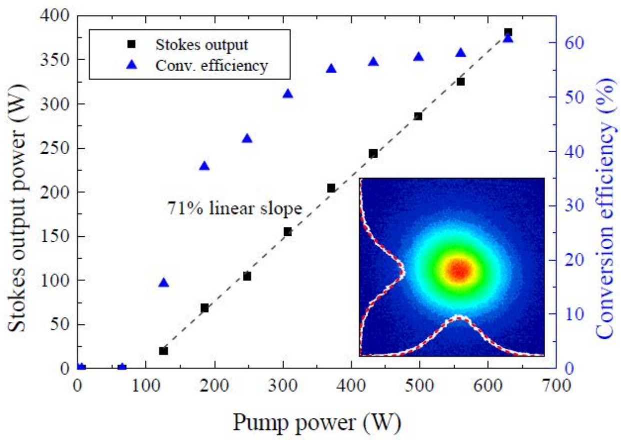


**Figure 2 Power characteristic and output beam profile for the 700W pumped DRL.**

The above results were obtained for burst durations up to 10 ms. To date, indefinite pulse durations have been attempted but have been limited to a few seconds due to feedback issues from the DRL into the pump laser. We are working to solve these. The lack of saturation, damage or beam quality degradation indicates that heating of the diamond or its coatings is a problem at the current levels. We conclude that the present DRL designs are suitable for conversion of pums of power up to 1 kW, with potential for higher pump powers depending on the detailed dynamics of phonon decay and its effect on the temperature profile in the diamond.

For further published details see:
A. McKay, O. Kitzler and R.P. Mildren, *Opt. Express*, vol. 22, 6707–6718 (2014).
R.J. Williams, A.M. McKay, O. Kitzler and R.P. Mildren, *Opt. Lett.*, vol. 39, 4152–4155 (2014)
R.J. Williams, J. Nold, M. Strecker, A. McKay, T. Schreiber and R.P. Mildren, Laser & Photon. Reviews, vol. 9, (accepted 11 Jun; 2015)

**2) Eye-safe DRLs**

There has been much development in pulsed eye-safe lasers due to their importance in field applications such as remote sensing and optical countermeasures. Optical parametric oscillators and solid-state lasers such as resonantly-pumped Er:YAG are currently favoured approaches, particularly for high pulse rate systems. However, the brightness of these systems is challenging to maintain when scaling output power. We have exploited Raman beam cleanup and the excellent thermal properties in diamond to demonstrate an approach for further scaling the brightness.

The system is based on a DRL providing second Stokes conversion of a 1064 nm Q-switched laser (see Figure 3a). A major focus of the investigation was to exploit Raman beam cleanup to provide high brightness output from a low beam quality source. An input beam pulsed at 36 kHz pulse repetition frequency and with power 40 W and a beam quality factor $M^2 = 3$–$4$ was used. The output beam (16 W at 40% conversion efficiency) had a quality factor of $M^2 = 1.17 \pm 0.08$ which is a factor of 2.7 times lower than that of the input beam, resulting in a much higher overall brightness (**540 MW/cm²/sr** cf. 320 MW/cm²/sr for the input beam). The output power, brightness, and brightness enhancement obtained represent significant advances in performance of Raman lasers as well as other competing kHz-pulsed eye-safe technologies (see Figure 3b).

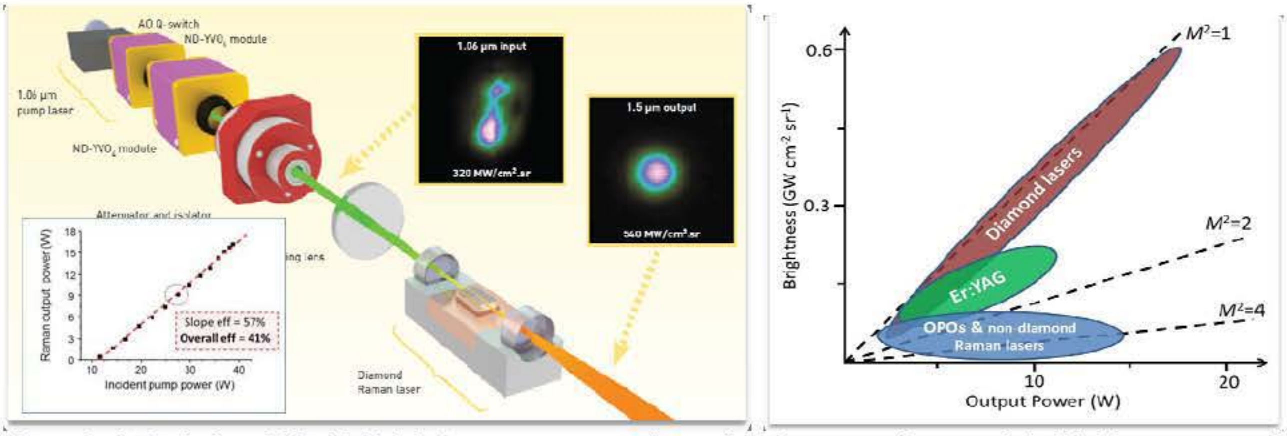


**Figure 3 a) Depiction of the DRL brightness converter along with the power characteristic. b) shows an overall comparison of brightness and power performance of DRLs compared to other major eye-safe pulsed laser technologies.**

The brightness enhancement (BE) is a product of the efficiency and the ratio of input and output beam qualities. For the first time the BE factor is shown to exceed unity for an external cavity Raman laser and is achieved using a compact device of length 2-3 cm. No thermal effects are observed in the power characteristic and beam quality at the current power levels, consistent with our models for thermally-induced lensing and birefringence. Based on the results above for cw lasers, diffraction limited beam powers above 0.4 kW are anticipated, a level which would correspond to brightness values above 10 GW/cm²/sr.

One potential issue for pulsed high power systems is crystal damage. During ns-pulsed laser characterization we observed occasional damage to the diamond. On two occasions, the diamond split in two along the beam axis in a {111} facet direction. The timing of the damage seemed to be spontaneous after an extended period of operation (tens of minutes) at relatively low output powers (less than 30% of the maximum power). As a result, we suspect that the mechanism is a result of interaction with point or line defects in the crystal that evolve in the intense beam zone, eventually leading to crystal cleavage along "soft" planes. Due to the low frequency of these events, we were unable to investigate the average power and peak power dependencies on the damage probability.

For further published details on this topic see:
A. McKay, O. Kitzler and R.P. Mildren, *Laser Physics Letters*, vol. 10, 105801 (2013)
A. McKay, O. Kitzler and R.P. Mildren, *Laser & Photon. Reviews*, vol. 8, L37-L41 (2014)

### 3) Fiber laser conversion

Nonlinear frequency conversion of fiber lasers is intrinsically challenging due to the comparatively low peak intensities generated in cw systems, and the typically-low spectral power density, unless steps are taken to narrow the output linewidth. Raman fiber lasers are progressing rapidly with substantial average output powers now demonstrated to generate wavelengths near 1178 nm for applications in medicine and laser guidestars. DRLs offer a bulk solid-state alternative with advantages of large wavelength jumps (1332 cm$^{-1}$ Stokes shift cf. <600 cm$^{-1}$ for Raman fibers), reduced line broadening (Raman linewidth is 1.5 cm$^{-1}$ cf. fibers >100 cm$^{-1}$) and without photo-darkening concerns.

We investigated two systems based on first Stokes conversion of seeded Yb-doped fiber amplifiers. The first was a DRL pumped using a 50 W cw fiber amplifier that was tunable in the range 1062-1068 nm (to produce Stokes output in the range 1237-1243 nm). The pump linewidth was 5 MHz (single longitudinal mode). The cavity and focus design was optimized for maximum efficiency at full pump power using the model of Sec 4 (see below). The laser threshold was 10 W and power increased to a maximum of **18.5 W** at the full incident power of 42 W (conversion efficiency of **44.9%**) – see Figure 4. The beam quality was diffraction limited as expected at the current power levels. Since the gain medium in the DRL is expected to be homogeneous and without spatial hole burning, there is potential to generate single longitudinal mode (SLM) output from a relatively simple system. Thus we also characterized the spectral composition of the output as a function of laser parameters. It was found that just above threshold it was possible to achieve SLM output under some conditions. However, at significant output powers, the output became highly multimode with linewidth of up to approximately 10 GHz. Future work will examine the line broadening mechanisms in detail with the aim of developing designs for efficient SLM Stokes generation. The topic of SLM Raman lasers has received very little attention in the literature to date in diamond or any other material. The combination of a tunable fiber laser with DRL wavelength conversion is a promising system for spectroscopy applications.

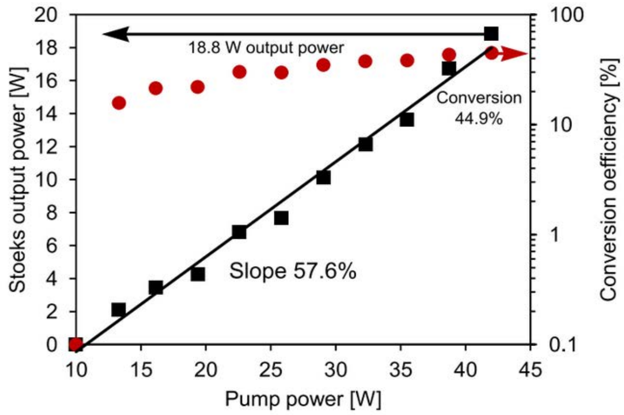


**Figure 4 DRL power and efficiency for a 50 W fiber laser pump.**

The second system was the 700 W fiber laser pumped DRL described in Sec 1. This work demonstrated output powers up to 380 W with a maximum conversion of 62% for burst periods substantially longer than the relaxation time for thermal gradients in the diamond. It shows that DRL technology is broadly applicable to high power fiber laser technologies with the caveat that the pump linewidth is not substantially larger than the Raman linewidth. As well as higher power and narrow linewidth output, future work of interest includes fiber laser pumped DRLs that target specific wavelengths for applications in remote sensing, guidestars, atom cooling and other highly wavelength specific applications. Future work will also investigate extension of the concept into the visible and UV, either by pumping directly with visible pump lasers, or through the use of intra-DRL-cavity harmonic generation.

For further published details see:
R.J. Williams, J. Nold, M. Strecker, O. Kitzler, A. McKay, T. Schreiber and R.P. Mildren, Laser & Photon. Reviews, vol. 9, (accepted 11 Jun; 2015)

### 4) Model development

The above empirical studies have been performed as functions of pump power with most cavity parameters fixed. It is important to better understand the importance of other cavity parameters on laser behaviour in order to assist with future optimization of designs. We thus developed a model to find laser parameters that maximize output power and efficiency as functions of pump power, crystal loss and optical design parameters. We have derived a new analytical solution to the Raman laser equations that is applicable to cw external cavity Raman lasers and form the basis for analyzing the high power DRLs.

A critical issue is found to be that up to 45% of the output Stokes power in earlier cw laser results is deposited as heat in the crystal, a large fraction of which is due to absorption loss. Thus the onset of thermal effects can be offset substantially by selecting operating conditions allowing high output coupling (thus lower intracavity fields) and short crystal lengths. Using elevated pump powers, although generally adding to the total heat load, enables higher optimum output couplings and thus higher efficiency in the presence of parasitic losses. Although it may be ultimately necessary to increase the pump and Stokes spot sizes to avoid damage, the model shows that the maximum intracavity Stokes intensity required to effectively deplete the pump beam (which is a necessary condition for efficient operation), is fixed for any pump power. Since the total intensity experienced by the diamond facets is primarily due to the resonant Stokes field, the system has the benefit of being scalable to higher powers without greatly increased risk of optical damage, as well as operating more efficiently.

The minimum DRL threshold observed to date is 10 W, yet there is interest in reducing this value to enable efficient devices at low power. Reductions in threshold require designs that can enhance the intracavity Stokes field, which is achieved by reducing cavity losses and output coupling and tighter focussing of the laser fields. Low loss diamond is therefore crucial. Reducing the pump spot size is relatively straightforward by using suitable focusing optics, however, keeping the Stokes field mode-matched is challenging in a near-concentric resonator. Folded cavities or cavities with internal lenses may be beneficial for supporting smaller mode waists. Using lowest loss available material, and for waist sizes approaching 10 μm, we expect thresholds as low as a few watts may be achievable. Waveguide structures represent a key approach to further reductions in laser threshold.

The analysis has assumed that the thermal lens induced in the diamond does not significantly change the pump and Stokes waist sizes. The thermo-optic effect is found to be the most likely primary cause for saturation at elevated power levels as a result of diamond's low susceptibility for other thermal lensing mechanisms that involve stress-optical effects (namely stress fracture, birefringence and end-facet distortion). Future work will need to involve these processes as DRL powers move towards the thermal regime. With a large number of applications near 1.5 μm, there is substantial interest in cw DRL converters to the second-Stokes wavelength of 1485 nm, or to even higher Stokes orders. By adding suitable equations for the cascade, the model is readily adaptable to accommodate higher order generation.

For further published details see:
O. Kitzler, A. McKay, D.J. Spence and R.P. Mildren, *Opt. Express,* Vol. 23, No. 7, 8590-8602, (2015)

**5) Additional Outcomes**

We also report on progress in three areas not originally planned in the proposal.

1) Raman beam combination

The technique of Raman beam combination, which has been investigated previously[4] for scaling the ***peak brightness*** of pulsed gas lasers (e.g., excimer lasers), automatically compensates for phase differences in each pump through the non-linear interaction, leading to a technique with fewer constraints on pump coherence compared to coherent beam combination techniques. Here we aim to use the technique in tandem with the outstanding thermal properties of diamond to scale the ***average brightness*** of pump lasers as well provide a wavelength shift. Laser systems of increased efficiency, brightness and extended wavelength range are critical to the next generation of defense systems, industrial material processing and for environmental sensing applications.

As a first step to test the concept, we investigated a three-beam Raman amplifier using mutually-incoherent nanosecond pump beams with kilowatt peak powers and seeded by a single input beam at the first Stokes. The pumps were brought together in a common focus inside a 9 mm-long diamond crystal, which, in the presence of the seed, were depleted by approximately 80% each at their peak (refer Figure 5). At the peak, 4.5 kW was transferred to the Stokes beam with a combiner efficiency of **72%** (optical input power to amplified output power). Such an efficiency is comparable with the maximum demonstrated for coherent beam combining approaches. To our knowledge, this work represents the first demonstration of Raman beam combining in any Raman crystal.

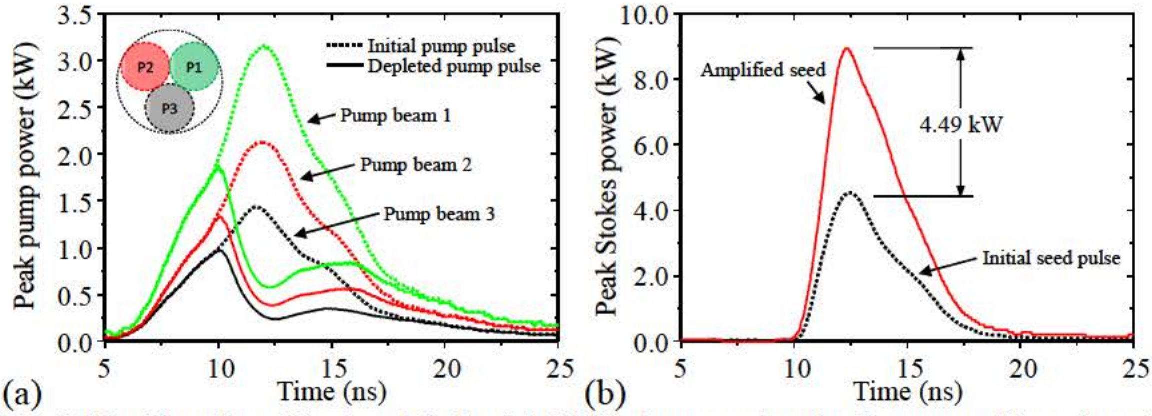


**Figure 5 a) Incident (dotted lines) and depleted (solid lines) pump pulses for three non-collinear input beams. b) Amplified (red solid) and seed pulse before the diamond beam combiner (black dotted line).**

The studies so far have been limited to 1 kHz pulse rate and 10 ns pulse durations. There is excellent potential for this technique to be extended to kilowatt average powers by increasing pulse rate and/or pulse duration. A critical issue will be the thermal limit for diamond which, as noted above, is presently not well understood. Future work is needed to understand pulse duration and power limits, as well as to optimize the combiner design in terms of crystal and beam geometries.

For further published details see:
A. McKay, D.J. Spence, D.W. Coutts and R.P. Mildren, *Proceeding of Advanced Solid-State Lasers*, Shanghai, Nov 16-21, ATu5A.1 (3 pages), (2014). Postdeadline paper

[4] M. Smith, D. Trainor, and C. Duzy, IEEE J. Quantum Electron. **26**, 942–949 (1990).

2) Evidence for SBS in diamond

Our studies into high power cw beam conversion has led to what we believe is the first reported observation of stimulated Brillouin scattering (SBS) in diamond. The high-Q external cavity architecture is amenable to generating Stokes intensities above 10 MW/cm$^2$ and in a regime for exploring other nonlinear effects. The output spectrum shown in Figure 6 reveals a secondary peak at a separation of 2.4 cm$^{-1}$ (72 GHz) from the long-wavelength-side of the main Stokes line. This separation value is consistent with backwards-generated SBS from the longitudinal acoustic mode in diamond. We noted that the amplitude decreased markedly when the DRL operated with a broader Stokes linewidth, consistent with the dependence of SBS gain on pump spectral power density. No secondary peaks in the pump spectrum in the vicinity of the main peak were evident (shown in inset to Fig), suggesting that the satellite is produced by SBS of the 1234 nm Raman line.

SBS may pose challenges for narrow-linewidth higher-power DRLs and may need to be managed by reducing the intra-cavity spectral power density via, for example, increasing output coupling or mode size, which both increase the threshold of the Raman laser. Future work is suggested to determine the gain coefficient and linewidth in order to more accurately predict SBS thresholds and impact on Raman laser dynamics. On the flipside, SBS in diamond may be of significant interest in the fields of microwave-photonics, narrow-linewidth lasers and Brillouin frequency-combs, as a medium with high SBS gain, high damage threshold, enhanced transparency range and high Brillouin frequency. Indeed we have shown that SBS can be obtained in a bulk crystal, in contrast to other crystalline media for which SBS has only been achieved in nano-structured devices, so that the effect may exploited across a greater range of platforms.

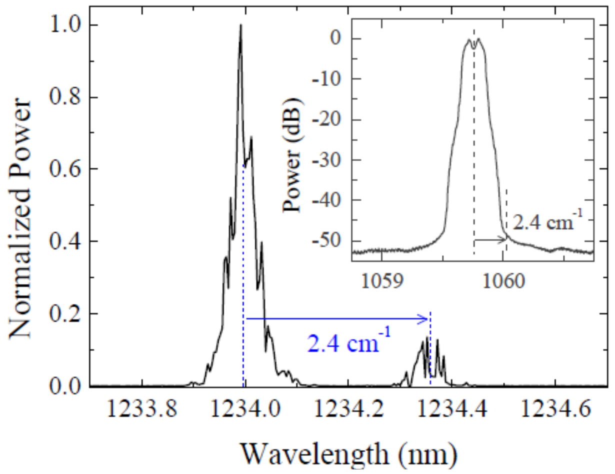


**Figure 6 Optical spectrum of a DRL pumped with 309 W at 0.16 nm linewidth showing the 2.4 cm$^{-1}$ shift from the Stokes line. The inset shows the spectrum of the pump laser (on a logarithmic power scale).**

For further published details see:
R.J. Williams, J. Nold, M. Strecker, O. Kitzler, A. McKay, T. Schreiber and R.P. Mildren, Laser & Photon. Reviews, vol. 9, (accepted 11 Jun; 2015)

3) Optical techniques for processing materials with a resolution less than the wavelength of light have advanced significantly in recent years. However, despite the highly selective nature of light-matter interactions, efforts to increase resolution to the atomic scale are hampered by rapid and efficient dissipation of the absorbed energy to the surrounding matrix. We have found that diamond surfaces exhibit the remarkable capability for patterning on ultra-deep sub-wavelength (<20 nm) length scales when exposed to intense ultraviolet light. Furthermore, the detailed shape of structures depends sensitively on the polarization of the ultraviolet beam with respect to lattice bond directions. The results have revealed that photons couple strongly to localized electronic states associated with the surface carbon-carbon bonds, potentially providing a mechanism for targeted single atom ejection. This research direction, which has emerged from high power laser investigations, has potential impact in other fields and is detailed in the publications listed below.

For further published details see:
C.G. Baldwin, J.E. Downes, C.J. McMahon, C. Bradac, and R.P. Mildren, *Phys. Rev. B*, vol. 89, 195422 (2014).

**Summary, Significance and Outlook:**
The project increased the highest power obtained for a diamond Raman laser by 15 times and up to 0.4 kW. A continuous wave diamond laser was demonstrated that generated 108 W at 1240 nm from a 320 W Nd:YAG pump laser. A modified design generating 380 W was demonstrated using a 630 W Yb-doped fiber laser system. In each case the performance was unsaturated and limited by the available pump power. The efficiencies and brightness achieved are found to be higher than expected by current theories for thermal effects in diamond. The project also developed a diamond laser capable of providing large increases in beam quality (from $M^2 = 3.5$ to $M^2 < 1.1$) and brightness (by 1.7 times), features that were used to demonstrate a pulsed eye-safe (1.5 μm) laser based on a Nd:YAG pump with an unprecedented brightness of 540 MW/cm$^2$/sr.

The project also made significant findings in the related areas of stimulated Brillouin scattering (the first evidence in diamond to our knowledge), beam combination in diamond and diamond laser-surface interactions.

The results are significant in showing that diamond Raman laser technology is an efficient wavelength converter for conventional high power laser technologies including Nd doped lasers and Yb-doped fiber lasers. Diamond's power handling capability now exceeds the highest achieved for any other Raman crystal by a factor of 22 and is competitive with Raman fibers. In addition, it is highly promising for brightness enhancement of beams and for beam combining of incoherent beams based on the technique of Raman beam combination. Compared to other nonlinear conversion schemes (such as harmonic generation in crystals and Raman fibers), diamond has important advantages of large wavelength Stokes shift, wavelength range (from UV to mid-IR) and suitability for conversion a variety of temporal domains (ultrafast to cw). It is thus found to be an outstanding nonlinear optical material in terms of many important categories of high power performance.

The findings in SBS and diamond-laser surface interactions are also significant. Solid-state SBS is presently of major interest for applications in on-chip microwave photonics and optical communications technologies. The high Brillouin frequency of diamond, in combination with its other extreme properties, holds promise in these applications for extending performance or solving shortcomings found in other materials. The findings in laser – surface interactions are significant for revealing an entirely novel laser-induced mass ejection mechanism and for potentially providing a new tool for creating diamond nano- and micro-devices.

The project raises several questions for further research.

As power scaling is a major motivation for studying diamond, one of the most important questions pertains to the extent to which laser power can be scaled before thermal effects become evident. The mechanism for heat deposition and its spatial dependence are found to be poorly understood. Thus further experiments with accompanying theoretical development are crucial to the development of accurate models.

The results on beam brightness enhancement and beam combination also reveal interesting directions for further research. The degree to which diamond lasers are able to generate high quality output from aberrated input beams, multimode input beams, or separate incoherent input beams (Raman beam combination) is not well understood. A more detailed knowledge in these areas is important for assessing the potential for diamond applications in areas such as directed energy.

The discovery of SBS in diamond, in bulk, opens up a range of new questions regarding its potential negative impacts on Raman laser performance and whether the can process be exploited in other areas of science and technology. Determination of the SBS gain coefficient and linewidth are areas of immediate focus that will support future model development.

Finally, there are a large number of questions relating to the findings on laser surface etching of diamond. This field is immature with major gaps in knowledge in the characteristics of etching, the underlying mechanism and its potential applications that require further investigation.

**List of Publications and Significant Collaborations that resulted from the AOARD supported project:**

*a) papers published in peer-reviewed journals*

R.P. Mildren, “Intrinsic optical properties of diamond,” in Optical Engineering of Diamond (Wiley-VCH) pp1-34, (2013) **(Book Chapter)**

R.P. Mildren, A. Sabella, O. Kitzler, D.J. Spence and A. McKay, “Diamond Raman laser design, performance and prospects,” in Optical Engineering of Diamond (Wiley-VCH), Chapter pp239-276, (2013) **(Book Chapter)**

A. McKay, O. Kitzler and R.P. Mildren, “An efficient 14.5W diamond Raman laser at high pulse repetition rate with first (1240 nm) and second (1485 nm) Stokes output,” Laser Physics Letters, vol. 10, 105801 (2013)

A. McKay, O. Kitzler and R.P. Mildren, “High power tungstate-crystal Raman laser operating in the strong thermal lensing regime,” Optics Express, vol. 22, 707-715, (2014)

A. McKay, O. Kitzler and R.P. Mildren, “Thermal lens evolution and compensation in a high power KGW Raman laser,” Optics Express, vol. 22, 6707–6718 (2014).

C.G. Baldwin, J.E. Downes, C.J. McMahon, C. Bradac, and R.P. Mildren, “Nano-structuring and oxidation of diamond by novel two-photon UV surface excitation: An XPS and NEXAFS study,” Phys. Rev. B, vol. 89, 195422 (2014).

R.J. Williams, A.M. McKay, O. Kitzler and R.P. Mildren, “Investigating diamond Raman lasers at the 100 W-level using quasi-cw pumping,” Opt. Lett., vol. 39, 4152–4155 (2014)

A. Lehmann, C. Bradac, and R. P. Mildren, “Two-photon polarization-selective etching of emergent nano-structures on diamond surfaces,” Nature Commun. vol. 5, 3341 (2014)

A. McKay, O. Kitzler and R.P. Mildren, “Simultaneous brightness enhancement and wavelength conversion to the eye-safe region in a high-power diamond Raman laser,” Laser and Photonics Reviews, vol. 8, L37-L41 (2014)

O. Kitzler, A. McKay, D.J. Spence and R.P. Mildren, "Modelling and Optimization of Continuous-Wave External Cavity Raman Lasers," Opt. Express, Vol. 23, No. 7, 8590-8602, (2015)

*b) papers published in non-peer-reviewed journals or in conference proceedings,*

R.P. Mildren, H.M. Pask, D.J. Spence, S.D. Jackson, D.W. Coutts and M.J. Withford, “Novel indigenous photonics technologies for defence,” Australian Defence Engineering Conference (ADEC), Tues 25th Nov. Paper 01, (2014).

A. McKay, D.J. Spence, D.W. Coutts and R.P. Mildren, “Non-collinear beam combining of kilowatt beams in a diamond Raman amplifier," Advanced Solid-State Lasers, Shanghai, Nov 16-21, ATu5A.1 (3 pages), (2014). *Postdeadline*

*c) conference presentations,*

O. Kitzler, A. McKay, R. P. Mildren, "Characterization of a Single-frequency-pumped Continuous-wave Extracavity Diamond Raman Laser", in CLEO Europe, OSA Technical Digest, (Optical Society of America, 2013), paper CD-P.37.

A. McKay, O. Kitzler, R. P. Mildren, "Power Scaling of Efficient Diamond Raman Lasers with 1240 nm and 1485 nm Output", in CLEO Europe, OSA Technical Digest, (Optical Society of America, 2013), paper CA-1.

O. Kitzler, A. McKay,V. M. Hadiya and R.P. Mildren, "Characterization and Optimization of External Cavity Continuous-wave Diamond Raman Lasers," Frontiers in Optics/Laser Science XXIX (FiO/LS) meeting. 6-10 October, Orlando, Florida. Paper LTh2H.1, (2013)

A. Sabella, J.A. Piper, R.P. Mildren, "Tuneable mid-infrared diamond Raman laser," Photonics West, San Francisco, Paper. [8959-12] Feb 1-6 (2014)

R.J. Williams, O. Kitzler, Aaron M. McKay, R.P Mildren, "Power scaling of diamond Raman lasers beyond 100 W using quasi-cw pumping,," Photonics West, San Francisco, Paper. [8964-17] Feb 1-6 (2014)

A.M. McKay, O. Kitzler, R.P. Mildren, "An analysis of thermal effects in high power (8 W) KGW Raman lasers," Photonics West, San Francisco, Paper. Photonics West, San Francisco, Paper. [8964-36] Feb 1-6 (2014)

A. Lehmann, C. Baldwin, J.E. Downes, R.P. Mildren, "Polarization selectable nano-pattern formation on diamond surfaces by 2-photon ultraviolet desorption," Photonics West, San Francisco, Paper. Photonics West, San Francisco, Paper [8968-23] Feb 1-6 (2014)

R.P. Mildren, "High Power Beam Conversion of Pulsed and Continuous Wave Lasers in Diamond," Int Conf on High Power Lasers and Applications, Aug 26-30, Chengdu China (2014) *Invited*

R.P. Mildren, "Development of UV Laser-induced Emergent Nano-structures on Diamond," Int Conf on High Power Lasers and Applications, Aug 26-30, Chengdu China (2014)

R. J. Williams, O. Kitzler, J. Nold, M. Strecker, A. McKay, T. Schreiber, and R.P. Mildren, "290 W fiber-laser-pumped diamond Raman laser," accepted for oral presentation at Europhoton (conference of the European Physical Society) Neuchatel, Switzerland (2014).

R.P. Mildren, "Characteristics of emergent nano-structures formed on diamond by two-photon UV etching," IEEE Photonics Conference, San Diego, Oct 12-16 (2014)

A. McKay, R.J. Williams, O. Kitzler, H. Jasbeer, S. Sarang and R.P. Mildren, "High Power Raman Beam Conversion in Synthetic Diamond," Advanced Solid-State Lasers, Shanghai, Nov 16-21, AM5A.53 (2014). *Invited*

A. McKay, D.J. Spence, D.W. Coutts and R.P. Mildren, "Non-collinear beam combining of kilowatt beams in a diamond Raman amplifier," Advanced Solid-State Lasers, Shanghai, Nov 16-21, ATu5A.1 (2014). *Postdeadline*

R.P. Mildren, "Emergent Nano-Structures Formed on Diamond by Two-Photon UV Etching" MRS Fall Meeting, L14.04, Boston, USA, Dec 4, (2014)

O. Kitzler, A. McKay, R.J. Williams, and R.P. Mildren, "Undoped Synthetic Diamond for Applications in Raman Beam Conversion," MRS Fall Meeting, R8.05 , Boston, USA, Dec 4, (2014)

C G Baldwin, J E Downes, C J McMahon, and R.P. Mildren, "Investigation of UV etched diamond surfaces using XPS and auger yield NEXAFS for extreme surface sensitivity", oral presentation at Australian Synchrotron User Meeting, Melb., Dec (2012)

R.P. Mildren "Nano-scale properties of UV-photo etched diamond surfaces: Evidence for photo-selective bond scission," OzCarbon Conference, University of Melbourne, Paper 1, p26, Dec 2-4 (2013).

R.J. Williams, O. Kitzler, A.M. McKay, and R.P. Mildren, "Planar diamond waveguide Raman laser operating at 573 nm and 620 nm," Australian & New Zealand Conference on Optics & Photonics (ANZCOP). 8 – 11 December, postdeadline paper (2013) *Postdeadline Paper*

O. Kitzler, A.M. McKay, R. Williams, V. M. Hadiya, and R. P. Mildren, "Fibre laser pumped continuous-wave external cavity diamond Raman laser," Australian & New Zealand Conference on Optics & Photonics (ANZCOP). 8 – 11 December (2013)

A.M. McKay, O. Kitzler and R. P. Mildren, "Strong astigmatic lensing in high-power (>7 W) tungstate external-cavity Raman lasers," Australian & New Zealand Conference on Optics & Photonics (ANZCOP). 8 – 11 December (2013)

R.J. Williams, O. Kitzler, A.M. McKay, and R. P. Mildren, "Quasi-CW-pumping of diamond Raman lasers," Australian & New Zealand Conference on Optics & Photonics (ANZCOP). 8 – 11 December (2013)

C.G. Baldwin, J E Downes, C J McMahon, C Bradac, and R.P. Mildren, "Evidence for atomic-scale nanostructuring of diamond surfaces via two-photon UV technique", accepted for oral presentation at International Conference on Nanoscience and Nanotechnology, Adelaide, Feb (2014)

R.P. Mildren, "Emergent nano-structuring on diamond by two-photon UV etching," Combined Australian Materials Societies (CAMS 2014), Nov 26-28 2014. *Invited Paper*

*d) manuscripts submitted but not yet published, and*

R.J. Williams, J. Nold, M. Strecker, O. Kitzler, A. McKay, T. Schreiber and R.P. Mildren, "Efficient Raman frequency conversion of high-power fiber lasers in diamond," Laser and Photonics Reviews, vol. 9, (accepted 11 Jun; 2015)

*e) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.*

Numerous industry visits have been made in the context of commercial aspects pertaining to this work. Mildren visited AFRL in Albuquerque in Feb 2014. Part of this work was performed in collaboration with the J. Nold, M. Strecker and T. Schreiber of the Fraunhofer Institute of Applied Optics and Precision Engineering, Jena, Germany.